COMPARISON OF MICROWAVE-ASSISTED HYDRODISTILLATION WITH THE CONVENTIONAL HYDRODISTILLATION METHOD IN THE EXTRACTION OF ESSENTIAL OIL (LEMONGRASS AND STAR ANISE)

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ABSTRACT

Microwave-assisted hydrodistillation (MAHD) has recently been developed for the extraction of essential oils from plant materials. In this study, microwave-assisted hydrodistillation was investigated for the extraction of essential oils from lemongrass and start anise and the results were compared with those of the conventional hydrodistillation in terms of extraction time, extraction yield/efficiency and chemical composition. Microwave-assisted hydrodistillation was efficient in extraction in terms of extraction time and energy saving. Lemongrass and star anise was in the ratio of 1:10 with water and the essential oils components were identified using GCMS. There were significant different in the extraction yield of essential oils from both of the method and higher yield were obtained from MAHD method. Results of analysis from gas chromatography-mass spectrometry indicated that the use of microwave in hydrodistillation did not adversely influence the composition of essential oils. Microwave-assisted hydrodistillation was found to be environmentally friendly due to its shorter extraction time and therefore lower energy consumption.

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ABSTRAK

Pergabungan microwave dengan penyulingan berasaskan air (MAHD) telah diaplikasikan dalam penghasilan minyak asli daripada tumbuhan sejak kebelakangan ini. Dalam kajian ini, MAHD digunakan untuk menghasilkan minyak asli daripada serai dan bunga lawang dan hasilnya dibandingkan dengan penyulingan tradisional berasaskan air (HD) dari segi jangka masa proses, kadar penghasilan dan komposisi minyak asli. MAHD efisien dalam penghasilan minyak asli dari segi jangka masa proses dan menjimatkan tenaga. Serai dan bunga lawang yang digunakan adalah dalam nisbah 1:10 dengan air dan komposisi minyak asli dikenalpasti dengan menggunakan menggunakan alat GC-MS. Perbezaan jangka masa proses yang ketara didapati daripada dua cara tersebut dan penghasilan minyak asli yang lebih tinggi didapati dalam cara MAHD. Data analisis yang diperoleh daripada GC-MS menunjukkan penggunaan microwave dalam penyulingan berasaskan air tidak mempengaruhi komposisi minyak asli. MAHD tidak memberi kesan negatif terhadap alam sekeliling kerana jangka masa proses yang rendah dan justeru penggunaan tenaga yang rendah.

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LIST OF ABBREVIATIONS

HD	=	Hydrodistillation
MAHD	=	Microwave-Assisted Hydrodistillation
GC-MS	=	Gas Chromatography-Mass Spectrometer
SFE	=	Supercritical Fluid Extraction
MAE	=	Microwave-Assisted Extraction
MASD	=	Microwave Accelerated Steam Distillation
MASE	=	Microwave-Assisted Solvent Extraction
SFME	=	Solvent-Free Microwave Extraction
MHG	=	Microwave Hydrodiffusion and Gravity
SPME	=	Solid Phase Micro Extraction

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LIST OF SYMBOLS

˚ C	=	Degree Celsius
%	=	Percentage
kPa	=	Kilo-Pascal
Hz	=	Hertz
MHz	=	Mega-Hertz
GHz	=	Giga-Hertz
W	=	Watts
mL	=	Mili-Liter
g	=	Grams
L	=	Liter
min	=	Minutes
hr	=	Hours
m	=	Meter
mm	=	Mili-Meter
µL	=	Micro-Liter
µm	=	Micro-Meter
mL/min	=	Mili-Liter Per Minute
w	=	Weight
w/w	=	Weight of Oil/Weight of Plant Materials

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

In food industry, the use of herbs and plants in the production of essential oils becomes significance because of their use in many applications including flavours and fragrances as well as in medicine. Essential oils contain the DNA of the plant or herb they are extracted from. They are complex mixtures of volatile compounds such as terpenes (mostly monoterpenes and sesquiterpenes), phenolics and alcohols (*Lucchesi et al., 2004*), which gives the characteristic odour and flavor closely associated with the vegetative matter they are obtained.

Essential oils can be isolated using a number of isolation methods such as hydrodistillation, steam distillation and organic solvent extraction. The conventional method for extraction of essential oils is hydrodistillation. In this method, a mixture of water and plant materials are heated and followed by liquefaction of the vapors in a condenser to evaporate the essential oils. However, this method resulted in several disadvantages including losses of volatile compounds and long extraction time (*Khajeh et al., 2004*). Recently, microwave-assisted hydrodistillation (MAHD) has gained attention and widely used to obtain essential oils from plant materials. Plant

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material placed in a Clevenger apparatus and heated inside microwave oven for a short period of time. In this study, MAHD was applied as a new technology for the extraction of essential oils from lemongrass and star anise.

1.2 Problem Statement

The worldwide market for essential oils due to its increasing importance in pharmaceutical, fragrances and food industry trigger the research on new techniques for a better extraction. Conventional hydrodistillation involves distillation of plant material in water for long period. Hydrodistillation is economically viable and safe and it is the most common method for extraction of essential oils. However, the market values of essential oils lead to the application of microwave energy in the extraction to obtain higher quality oils and effective extraction.

Conventional hydrodistillation method is time consuming and low efficiency. In conventional hydrodistillation, heat transfer depends on thermal conductivity. Heat is transferred from the heating medium to the interior of the sample (*Bousbia et al., 2009*) resulted in the slowly increase in temperature. In microwave-assisted hydrodistillation, microwaves are volumetrically distributed (*Bousbia et al., 2009*). Due to the volumetric heating effect, a faster increase in temperature can be obtained depending on the microwave power and the dielectric loss factor of the material being irradiated. Extraction of essential oils from plant materials started right after the sample achieved boiling point. This causes an important difference in extraction time between the conventional and microwave-assisted hydrodistillation.

Instead of extraction time, losses of volatile compounds are another problem that arises when using conventional hydrodistillation method. In microwave-assisted hydrodistillation heat energy is produced by microwave energy. The efficiency of

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MAHD depends strongly on the dielectric constant of water and the matrix (*Brachet et al., 2002*). It caused the rapid delivery of energy to the total volume of solvent/sample in which the sample reaches its boiling point rapidly. Heat is originated through the molecular motions (*Brachet et al., 2002*) and the rise in temperature within the plant cells is similar to that occurring outside the cells. The external cell walls break apart once the pressure within the glands reaches certain level (*Chemat et al., 2005*) to release the essential oil. The rapid heating of plant materials by the microwave energy minimizes the losses of volatile compounds from the plant materials.

1.3 Objectives

The objective of this project is:

i) Using a new technology, the microwave-assisted hydrodistillation as an alternative method to extract essential oil from plant materials.

ii) Extract essential oil from plant materials using conventional hydrodistillation method.

iii) Analyze the overall performances of microwave-assisted hydrodistillation and make a comparison between microwave-assisted hydrodistillation and conventional hydrodistillation method.

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1.4 Scope of study

There are some important tasks to be carried out in order to achieve the objectives of this study. The important scopes have been identified and all the research works will be base on the scopes throughout the study.

i) In this study, we have been restricted the raw materials (Lemongrass and Star Anise). The extraction of essential oil will be carried out on these raw materials using conventional hydrodistillation and microwave-assisted hydrodistillation.

ii) Analysis on the essential oil will be carried out using Gas Chromatography – Mass Spectrometer (GC-MS) to determine the components of the essential oil for particular raw material.

iii) The comparison on both of the method will be in terms of :

- a) Extraction time
- b) Extraction yield/efficiency
- c) Chemical composition
- d) Cost of operation

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1.5 Rationale and Significance

The rationale and significance of this study is:

i) The market value of essential oils increases due to its importance in pharmaceutical, fragrances, and food industry. There is a need to explore new technique on extraction to replace conventional extraction method to get a better extraction in terms of time, cost, and quality of essential oils.

ii) Extraction of essential oil using microwave-assisted hydrodistillation involved short extraction time, high extraction efficiency, and minimize the losses of volatile compounds in the plant materials.

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CHAPTER 2

LITERATURE REVIEW

2.1 Essential Oils

Essential oils are the volatile fraction of the secondary metabolites produced plants (*Ramanadhan et al., 2005*). The essential oils extracted from the plant materials contain the DNA of the plant. It is normally very concentrated that it gives 100 times the flavoring strength of the parent plant (*Mohamed, 2005*). Essential oils bearing plants have their value in food industry, fragrance and pharmaceutical.

Essential oils are highly complex compounds and their constituents included oxygenated compounds. They are a group of natural organic compounds that are predominantly composed of terpenes (hydrocarbons) and terpenoids (oxygen containing hydrocarbons). Essential oils also contain simple phenols, sulphur containing mustard oils, methyl anthranilate and coumarins. Majority of them are fairly stable and soluble in high strength alcohol but have poor water solubility.

Terpenes and terpenoids in the plant were built from the basic 3-methyl-3-butenyl pyrophosphate. The 5-carbon unit of this molecule is the source of the

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isoprene unit, and combination two of these units give rise to geranyl pyrophosphate to form the skeleton of monoterpenes (10 carbons). Subsequently combination of 3 of these units gives rise to farnesyl pyrophosphate to form the skeleton of the sesquiterpenes (15 carbons). These complex mixtures of volatile compounds give the characteristic odour and flavor associated with the vegetative matter.

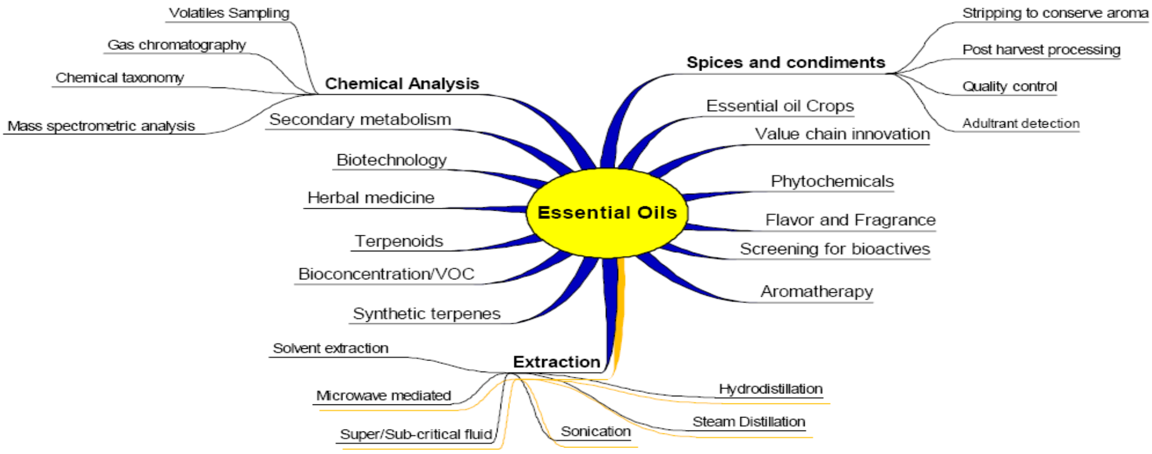

Figure 2.1: Tree diagram showing the wide branching of specializations in the field of essential oils (*Ramanadhan et al., 2005*)

In order to obtain the essentials oils from the plant materials, the isolation by physical means have to be carried out. The physical methods are direct distillation of essential oils, water steam distillation of essential oils or organic solvent extraction of organic compounds. The conventional method for the extraction of essential oils such as Soxhlet extraction method, liquid-liquid, and solid-liquid extraction are characterized by lengthy extraction procedures, consumption of large amount of solvent and energy and losses of some volatile compounds.

In the recent years, several studies have been conducted on new techniques to extract essential oils from plant materials. Among the techniques introduced was ultrasonic extraction, supercritical fluid extraction (SFE), extraction with subcritical or critical water, and application of microwave technique such as microwave-assisted extraction (MAE), solvent-free microwave extraction (SFME), microwave accelerated steam distillation, microwave hydrodiffusion and gravity (MHG), and

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microwave-assisted hydrodistillation. These new techniques is proved to be effective in the extraction of essential oils in which they involved shorter extraction time compared with the conventional method, higher yield and better quality of essential oils, minimize the consumption of solvent, energy saving and therefore environmentally friendly.

Table 2.1: Important essential oils (*Gunther, 1994*)

Name of oil	Method of production	Part of plant used
Almond	Steam distillation	Kernels
Bay	Steam distillation	Leaves
Bergamot	Expression	Peel
Caraway	Steam distillation	Seed
Cassia	Steam distillation	Leaves and twigs
Cedarwood	Steam distillation	Red core wood
Cinnamon	Steam distillation	Bark
Citronella	Steam distillation	Grass
Clove	Steam distillation	Buds
Coriander	Steam distillation	Fruits
Eucalyptus	Steam distillation	Leaves
Geranium	Steam distillation	Leaves
Jasmine	Cold pomade	Flowers
Lavender	Hydro-distillation	Flowers
Lemon	Expression	Peel
Orange	Expression, distillation	Peel
Peppermint	Steam distillation	Leaves and tops
Rose	Steam distillation, solvent, enfleurage	Flowers
Sandalwood	Steam distillation	Wood
Spearmint	Steam distillation	Leaves
Tuberose	Solvent, enfleurage	Flowers
Wintergreen	Steam distillation	Leaves
Ylang-ylang	Steam distillation, solvent extraction	Flowers

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2.2 Plant Materials

2.2.1 Agarwood

Agarwood (Gaharu) is a dark resinous heartwood that forms in *Aquilaria* trees when they become infected with a type of mold. There are 25 species of *Aquilaria* and 15 species are reported to form agarwood (*Barden et al., 2002*). In the Malaysia forests, the main species producing agarwood is *A. malaccensis* as it is commonly known (*Nor Azah et al., 2008*).

Agarwood is the resin impregnated, fragrant and highly valuable heartwood found in species of *Aquilaria*. It ranks among the most highly valuable traded forest products world-wide (*Wollenberg, 2001*). The wood released fragrance that is considered as scent when it is burnt.

Formation of agarwood occurs in the trunk and roots of trees that have been infected by a dematiaceous (dark-walled) fungus. As a response, the tree produces a resin high in volatile organic compounds to suppress or retarding the fungal growth. The resin dramatically increases the mass and the density of the affected wood, while the unaffected wood of the tree is relatively light in colour. The affected wood changing its colour to dark brown or black.

A common method in artificial forestry is to inoculate all the trees with the fungus. High quality resin produced when a tree's natural immune response to fungal attack. Agarwood from this process is commonly regarded as first quality agarwood. When trees are deliberately wounded, leaving them more susceptible to a fungal attack to create an inferior resin, it is commonly called second quality agarwood.

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Agarwood oil made from the agarwood is very tenacious and the colour of the oils may vary from greenish brown to dark reddish brown. The tiniest drops is needed to fill the air with its soul evoking aroma. Regarding the distinctive fragrance, it is used as perfumes, an essential oil and aroma therapy. Agarwood oil consists of complex mixtures such as sesquiterpene, hydrocarbons, sesquiteterpene alcohols, and aliphatic hydrocarbons in which to be identified using Gas Chromatography-Mass Spectrometer (GC-MS).

Table 2.2: Chemical compounds in Malaysia Agarwood oils (*Nor Azah et al., 2008*).

Chemical compounds	RI	Selangor (%)	Kelantan (%)	Pahang (%)	Terengganu (%)
3-phenyl-2-butanone	1249	1.50	5.77	7.80	0.79
α-guaiene	1448	-	0.67	-	-
β-agarofuran	1477	1.69	1.98	0.69	0.50
α-agarofuran	1553	4.83	2.96	1.48	1.57
Nor-ketoagarofuran	1557	2.09	-	-	-
10-epi-γ-eudesmol	1618	11.54	9.03	8.10	3.32
Agarospirol	1631	14.86	5.49	7.11	18.86
β-eudesmol	1649	-	-	-	5.74
Jinkoh-eremol	1650	10.62	7.70	6.31	-
kusunol	1659	18.94	-	-	-
Jinkohol II	1751	4.71	-	-	-

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2.2.2 Lemongrass

Lemon grasses (*Cymbopogon Citratus*) are a group of commercially important tropical grasses. The leaves of lemon grasses contain up to 1.5 % essential oils with a typical lemon-like aroma (*Lewinsohn et al., 1997*). Lemon grasses are indigenous in tropical and semi-tropical areas of Asia, and are cultivated in South and Central America, Africa and other tropical countries (*Weiss, 1997*).

Lemon grass is a perennial fast-growing aromatic grass, growing to about 1 meter high with long and thin leaves. It produces a network of roots and rootlets that rapidly exhausted the soil. The main chemical components of lemon grass oil are myrene, citronellal, geranyl acetate, nerol, geraniol, nearl and traces of limonene and citral. Citral is the name given to a natural mixture of two isomeric acyclic monoterpene aldehydes, geranial (trans-citral, citral A) and neral (cis-citral, citral B) (*Lewinsohn et al., 1997*).

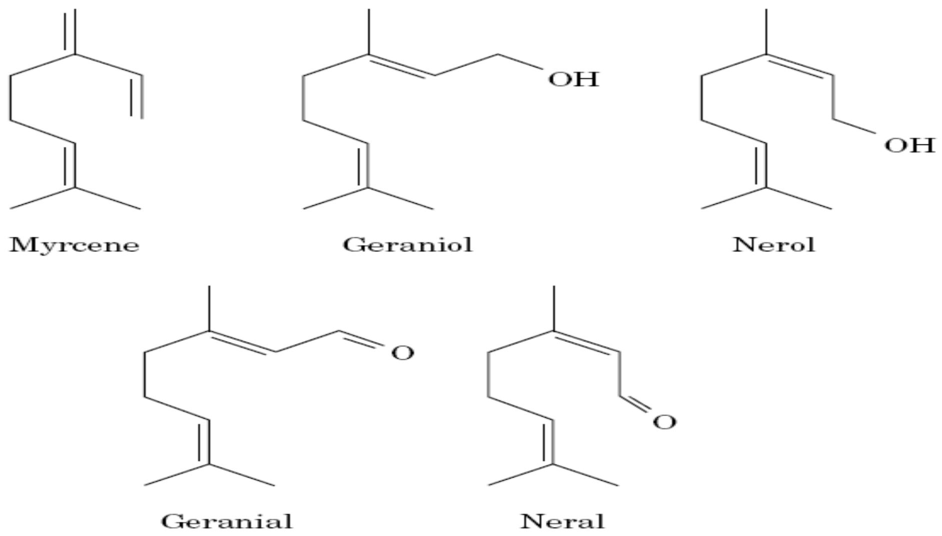

Figure 2.2: Chemical structure of the major constituents of lemongrass essential oil (*Lewinsohn et al., 1997*).

The medicinal part of lemon grass is the leaves, in which the lemon grass oil is extracted from the fresh or partly dried leaves by distillation. Lemon grass oil has a

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lemony, sweet smell and is dark yellow to amber and reddish in colour. Lemongrass oil can irritate a sensitive skin, so care should be taken. It is used in soaps, cosmetics, perfumes flavours and pharmaceutical products.

Table 2.3: Chemical constituents of Lemongrass oil determined by GC-MS (*Chimmalee et al., n.d.*)

Compound no.	Retention time (minute)	Compound	Relative amount (%)
1	5.28	Linalool	2.78
2	6.08	Citronellal	1.10
3	6.25	Verbenol	3.42
5	6.36	α-phellandren-8-ol	0.23
6	6.51	trans-Caran, 4, 5,	6.06
7	6.94	epoxi	0.33
8	7.17	cis-Caveol	0.96
9	7.21	β-Citronellol	0.32
10	7.41	cis-Geraniol	45.75
11	7.82	β-Citral	30.42
12	8.22	α-Citral	0.12
13	9.32	Geranyl formate	4.34
14	9.96	Geranyl acetate	0.37
15	12.45	trans-Caryophyllene	0.59
		Junipercamphor	2.83
		Unknown	

2.2.3 Star Anise

Star anise is defined as the dried, star-shaped multiple fruit of the tree of Illicium verum Hook., which is a member of the magnolia family (Magnoliaceae). Star anise fruits are produced on a medium-sized evergreen tree (Illicium verum

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